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Compound-specific stable carbon isotope values of fatty acids in modern aquatic and terrestrial animals from the Baltic Sea and Finland as an aid to interpretations of the origins of organic residues preserved in archaeological pottery

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The compound-specific stable carbon isotope ($\delta^{13}C$) analysis of organic residues is commonly used in the identification of lipid residues preserved in archaeological cooking vessels. This paper reports on the $\delta^{13}C$ values for saturated fatty acids from modern freshwater and brackish fish, wild mammals, domesticated animal muscle tissues, and their milk, collected from Finland and the Baltic Sea. Differences are shown to exist in the $\delta^{13}C$ values of carcass fatty acids of reindeer *(Rangifer tarandus)*, wild forest reindeer *(Rangifer tarandus fennicus)*, and other ruminant animals. Thus, the results reported in this paper show the importance of using modern reference fat data from similar ecological conditions to those of the studied archaeological site. This is especially vital at sites where wild fauna are known to have contributed significantly to human diet. In addition, discussion on problems related to representativeness of modern reference animal tissues for the interpretation of ancient fats is also carried out.

Keywords: Baltic Sea, Finland, compound-specific stable carbon isotopes, animal tissues, lipids, *n*-alkanoic acids

Introduction

Prehistoric hunter-gatherer-fisher populations lived by the coast of the Baltic Sea for millennia, and even farming communities in the area used the Baltic as a major food source. The Baltic Sea continued to provide an important part of the diet during medieval and historical periods (see e.g. Siiräinen 1981; Mannermaa 2016; Pääkkönen et al. 2016). The Baltic Sea is the second largest brackish basin in the world; including Kattegatt, its surface area is 415,000 km^2. The drainage area includes 14 countries and covers 1.74 million km^2. The catchment area of the Baltic Sea includes nearly 10,000 lakes that have a surface area > 1 km^2. Moreover, water salinity and the ecosystems vary markedly throughout the Baltic. The Baltic Sea is strongly influenced by large scale atmospheric circulation, the riverine freshwater supply, and the restricted salty water exchange coming through the Danish straits (see e.g. Gustafsson & Westman 2002:2; Omstedt et al. 2009:871). This results in more saline water in the south-western Baltic than in the Gulf of Bothnia and the Gulf of Finland (Fig. 1; HELCOM 2007:10). Raised river inflow in spring and summer, and the high salinity water flow during the autumn and winter from the North Sea into the Baltic Sea have

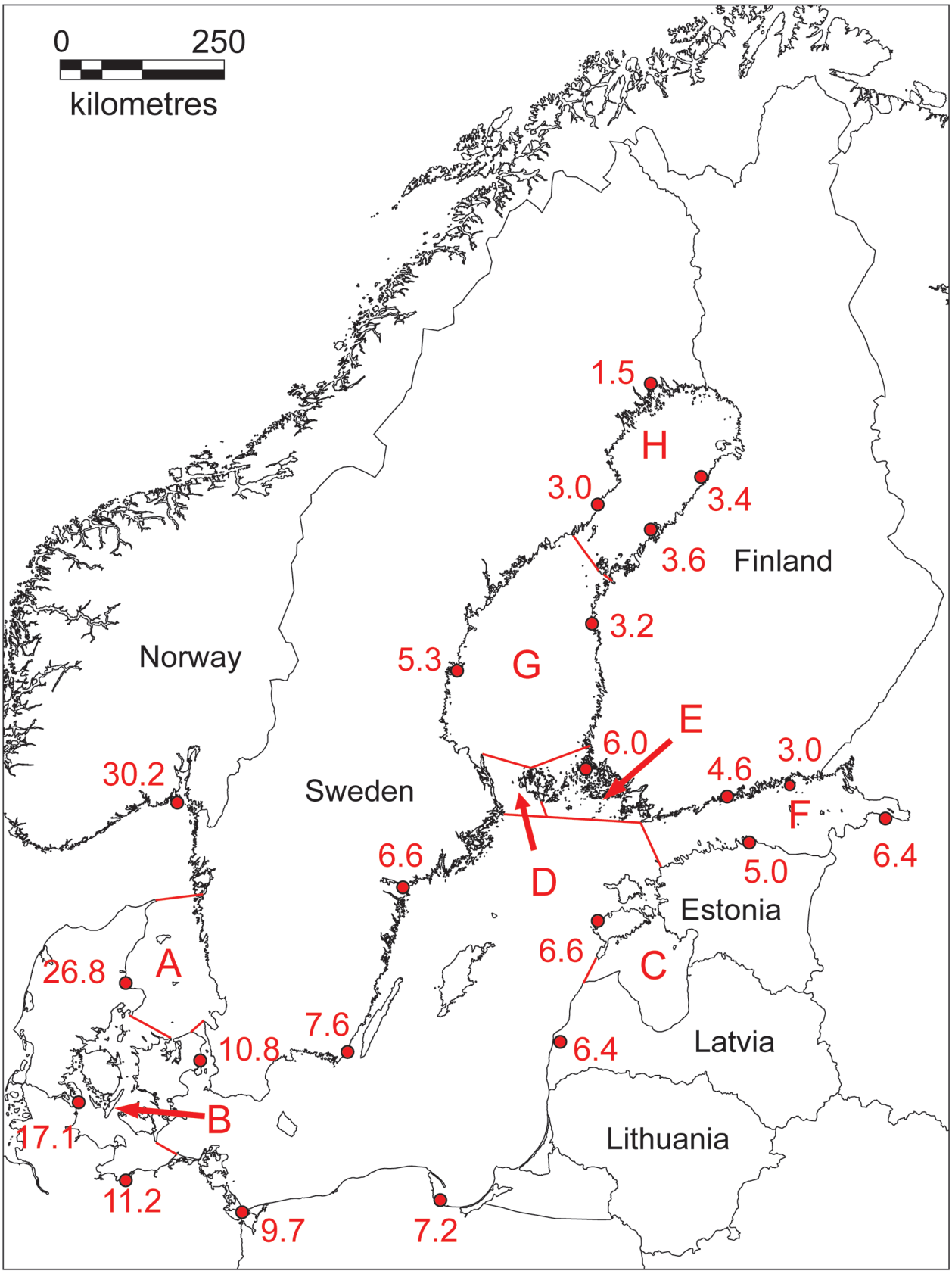

Figure 1. Study area, showing the sub-basins of the Baltic Sea referred to in the text. (A) Kattegatt, (B) Belt Sea, (C) Gulf of Riga, (D) Sea of Åland, (E) Finnish Archipelago Sea, (F) Gulf of Finland, (G) Sea of Bothnia, (H) Gulf of Bothnia. The salinity of the basin varies along the coastline with higher salinities in Kattegatt. The values 1.5–30.2 indicate the salinity (in ppt). Salinity data taken from DeFaveri et al. (2013:2533–2534).

important roles in the seasonal CO_2 cycle in the surface water. Furthermore, the Baltic Sea is also strongly influenced by the large riverine and atmospheric input of carbonaceous material from terrestrial and anthropogenic sources (Thomas & Schneider 1999:61, 65). A diverse range of fish species live in the Baltic Sea; in Finnish waters, 67 fish and two lamprey species can be found annually – and of these species more than 30% have both freshwater and brackish populations; some are also anadromous species (Urho & Lehtonen 2008:4).

Stable isotopic techniques are being used increasingly to study palaeoecology, ancient diet, and the wider aspects of food procurement in archaeological research. Such studies include the investigations of organic residues absorbed in the walls of ceramic vessels. The $\delta^{13}C$ values of palmitic ($C_{16:0}$) and stearic ($C_{18:0}$) acids of modern animals are recognised as essential aid for the identification of the origins of archaeological fats (Dudd & Evershed 1998; Evershed et al. 2002). Furthermore, the $\Delta^{13}C$ ($\delta^{13}C_{18:0}$ – $\delta^{13}C_{16:0}$) proxy is used to identify ruminant, porcine (non-ruminant), and dairy fats (Evershed et al. 1999; Copley et al. 2003). In addition to the aforementioned approaches, bulk $\delta^{13}C$ and $\delta^{15}N$ values can also be used to study diet. These bulk isotopes have, for example, been used when analysing human remains or charred food deposits from pottery (e.g. Lidén & Nelson 1994; Craig et al. 2007). Diet can also be examined by studying ancient human and faunal remains, with the help of compound-specific isotopes of amino acids (Webb et al. 2015). However, these techniques require either visible remains of charred food crusts or well-preserved skeletal remains. At Finnish prehistoric sites the bone preservation is often poor and usually only burnt bone fragments survive. Hence, absorbed organic residues offer an important archive, and the complex nature of the palaeoecosystem requires building a database of $\delta^{13}C$ values of *n*-alkanoic acids of modern reference organisms from the region.

The $\delta^{13}C$ values of the lipids from archaeological pottery in the Baltic area (see e.g. Craig et al. 2011; Cramp et al. 2014; Heron et al. 2015; Papakosta et al. 2015; Oras et al. 2017) have typically been compared to the modern isotope values gathered from non-local sources, i.e. the UK, Africa, the U.S. and Eurasian Steppe. This approach is acceptable when domesticates are considered, as the $\Delta^{13}C$ proxy effectively removes environmental influences and accentuates metabolic differences. However, interpretation becomes more complex when a hunter-gatherer-fisher diet is considered, as this can include a potentially more complex range of dietary resources. The situation is complicated further in the Baltic region due to the varying salinity of the basin. Furthermore, the northern latitudes present different ecological conditions for terrestrial animals which fundamentally affect their diets. For example, animals of the genus *Rangifer* have a different diet to ruminant animals living in more southerly latitudes. Due to this spatial variability and the unique nature of the Baltic Sea, it is important to further investigate the impact of these various environmental factors on the $\delta^{13}C$ values of modern mammal and fish muscle tissues from the northern Baltic region. Previous studies of Neolithisation in temperate Europe have necessarily focused on the key domesticates comprising the so-called "Neolithic package". However, the late arrival of animal husbandry and continued reliance on wild resource in the Baltic region creates a need for an expanded reference database of wild mammals and fish for use in identifying their potential contributions to lipid residues preserved in the pottery of hunter-gatherer-fisher and farming communities. Herein, we report the results of lipids analyses, with a particular focus on determination of fatty acids $\delta^{13}C$ values in fish and mammal muscle tissues, and the milk samples of domestic ruminants, collected from Finland and the Baltic Sea. The results are discussed in terms of their: (i) geographical origins, i.e. aquatic versus terrestrial; (ii) differences for wild species versus domesticated animals; and (iii) dietary differences, particularly for domesticated animals.

Materials and methods

The fish muscle tissues were sub-sampled from specimens acquired from the Natural Resources Institute Finland, the Archipelago Research Institute (University of Turku), recreational fishers, or purchased from fishmongers. The most common fish species currently living in the Baltic Sea and in freshwater systems in Finland were chosen for this study. The fish from the Baltic were mainly caught from the Finnish Archipelago Sea, the Sea of Åland, and the Sea of Bothnia. The freshwater fish were caught in the inland waterways of Central and Eastern Finland. None of the studied freshwater fish is a migratory species. Thus, all of the freshwater fish gathered were free from carbon isotope influences from the Baltic Sea, which could affect migratory species.

The ecological specimens of wild mammals were sub-sampled from the collections of the Natural Resources Institute Finland, the Zoological Museum of the University of Oulu, or obtained from recreational

hunters and a taxidermist. The milk samples were collected from organic cattle and goat farmers, and the samples of organic pork and beef were purchased from local supermarkets at different times during the summer and autumn of 2014. The oldest samples of wild mammals came from animals killed in the early 1980s, while the most recent samples were from 2006. The grey seals (*Halichoerus grypus*) were hunted in 2012–2014. All samples were stored in a deep freeze (–20°C) prior to analysis.

A total of 56 muscle tissue samples of fish from freshwater systems and the Baltic Sea, and 47 terrestrial and semi-aquatic mammal muscle tissue, and milk samples from Finland, and five seal samples from the Baltic Sea were studied (Table 1). Each sample was analysed using high-temperature gas chromatography (HTGC), GC-mass spectrometry (GC-MS) and GC-combustion-isotope ratio MS (GC-C-IRMS). The $\delta^{13}C$ values of fatty acid methyl esters (FAMEs) were corrected for the exogenous carbon added in the methylation process using the following mass balance equation (Rieley 1994):

$$\delta^{13}C_{FA}=\frac{\left((n+1)\times\delta^{13}C_{FAME}\right)-\delta^{13}C_{MeOH}}{n}$$

where: $\delta^{13}C_{FA}$ is the corrected $\delta^{13}C$ value of the fatty acid in ‰, $\delta^{13}C_{FAME}$ is the measured $\delta^{13}C$ value of the FAME in ‰, $\delta^{13}C_{MeOH}$ is the correction factor for the methanol used as derivatising agent, and n is the carbon chain length of the fatty acid.

The fossil fuel burning has led to an increase of ^{12}C in the atmosphere. Thus, in order to be able to compare modern and ancient stable carbon isotope values, the modern values have to be corrected for fossil fuel burning (Evershed et al. 1994:913). The decrease of $^{13}C/^{12}C$-ratio in the atmosphere is also transmitted to oceans via gas exchange at the air-sea interface. Freshwater reservoir effects can also alter the $\delta^{13}C$ values obtained from lakes and rivers, and riverine runoff to the Baltic will also have an effect to the $\delta^{13}C$ values of the organisms living in these water bodies (e.g. Lougheed et al. 2013; Philippsen 2013). Thus, further correction may be required for $\delta^{13}C$ values obtained from reference aquatic organisms before they can be used in archaeological or palaeoecological investigations. However, in this paper we present $\delta^{13}C$ values which have not been corrected for the aforementioned effects. This is done in order to provide reference $\delta^{13}C$ values that can be used for purposes other than archaeological investigations. In addition, by using the $\Delta^{13}C$ proxy any environmental effects of the type discussed above are removed, as the $C_{16:0}$ and $C_{18:0}$ fatty acids are affected equally.

Methanolic acid extraction

The muscle tissues of fish and mammals were sampled with a scalpel and freeze dried. The tissues were extracted using a modified direct methanolic acid extraction (Correa-Ascencio & Evershed 2014). Briefly, 5 ml of 5% *(v/v)* $MeOH/H_2SO_4$ was added to 0.02 g of the powdered tissue; samples were then heated at 70°C for 2 h, then 2 ml of H_2O (double-distilled, DCM extracted) added to quench the reaction. The supernatant was transferred to a clean culture tube and then extracted with *n*-hexane (3×3 ml). After blowing down under a stream of N_2, the extract was re-dissolved in *n*-hexane and an aliquot of the extract was trimethylsilylated with *N,O*-bis(trimethylsilyl) trifluoroacetamide (BSTFA; 20 µl) in readiness for the GC, GC-MS, and GC-C-IRMS analyses.

High temperature gas chromatography (HTGC)

HTGC analyses were performed using an Agilent Technologies 7890A GC. Helium was the carrier gas and a flame ionisation detector (FID) was used to monitor the column effluent. Diluted samples were introduced via on-column injection. The column was 15 m × 0.32 mm i.d. coated with dimethyl polysiloxane (Agilent Technologies, DB-1ht, film thickness, 0.10 µm). The temperature programming was from 50°C (hold 1 min) to 350°C at 10°C/min, followed by an isothermal hold at 350°C for 10 min. The FID was set to 350°C. Helium was used as the carrier gas and maintained at a constant flow of 4.6 ml/min. The eluting compounds were identified initially by their retention times.

Gas chromatography-mass spectrometry (GC-MS)

The GC-MS analyses of FAMEs were performed using a Finnigan Trace MS quadrupole mass spectrometer coupled to a Trace GC. Diluted samples were introduced using a PTV injector operating in the splitless mode onto a VF-23ms 60 m × 0.32 mm i.d. capillary column coated with cyanopropyl polysiloxane (film thickness, 0.15 µm, Agilent Technologies). The GC oven temperature was programmed from 50°C, following an isothermal hold for 2 min, to 250°C at 10°C/min, followed by anisothermal hold at 250°C for 10 min. The MS was operated in the electron ionisation (EI) mode (70 eV, emission current 149 µA)

Table 1. The $\delta^{13}C$ and $\Delta^{13}C$ ($\delta^{13}C_{18:0} - \delta^{13}C_{16:0}$) values of analysed muscle tissue and milk. All materials were from Finland unless stated otherwise.

	Lab code	$\delta^{13}C_{16:0}$ (‰)	$\delta^{13}C_{18:0}$ (‰)	$\Delta^{13}C$ (‰)	Place of catchment	Other remarks
Domesticated animals and milk						
Cattle (*Bos taurus*)	MM110	-29.5	-30.9	-1.4	Kärkölä	
Cattle (*Bos taurus*)	MM113	-28.8	-31.3	-2.4	Koski as.	
Cattle (*Bos taurus*)	MM117B	-29.6	-31.6	-1.9	n/a	
Cattle (*Bos taurus*)	MM111	-29.1	-30.6	-1.5	n/a	
Pig (*Sus scrofa domesticus*)	MM112B	-29.2	-27.6	1.6	n/a	
Pig (*Sus scrofa domesticus*)	MM114B	-30.0	-28.5	1.5	n/a	
Pig (*Sus scrofa domesticus*)	MM115	-29.3	-28.7	0.6	n/a	
Pig (*Sus scrofa domesticus*)	MM116B	-29.0	-27.5	1.5	n/a	
Milk (Northern Finncattle)	C21	-29.2	-32.8	-3.6	Laitila	
Milk (Northern Finncattle)	ML104	-28.9	-32.8	-3.9	Laitila	
Milk (Northern Finncattle)	ML105	-28.9	-33.6	-4.8	Laitila	
Milk (Northern Finncattle)	ML107	-28.7	-33.3	-4.6	Laitila	
Milk (Northern Finncattle)	ML108	-29.6	-33.7	-4.1	n/a	
Milk (goat, *Capra hircus*)	ML100	-27.5	-31.7	-4.3	Nauvo	Possible C_4 contamination?
Milk (goat, *Capra hircus*)	ML101	-29.3	-33.0	-3.7	Laitila	
Milk (goat, *Capra hircus*)	ML102	-29.3	-34.4	-5.1	Laitila	
Milk (goat, *Capra hircus*)	ML103	-29.7	-33.6	-3.9	Laitila	
Wild mammals						
Brown bear (*Ursus arctos*)	M10	-29.8	-30.0	-0.2	Ilomantsi	
Brown bear (*Ursus arctos*)	M4	-30.3	-30.3	0.1	Eno	Cub
Brown bear (*Ursus arctos*)	M7	-30.4	-29.9	0.5	Ilomantsi	
Brown bear (*Ursus arctos*)	MM134	-31.2	-30.8	0.4	Ilomantsi	
Brown bear (*Ursus arctos*)	M61	-28.1	-29.2	-1.0	Ilomantsi	Shot in oatfield
Eurasian beaver (*Castor fiber*)	M31	-30.3	-30.6	-0.3	Pori	
Eurasian beaver (*Castor fiber*)	M53	-31.9	-33.0	-1.1	Satakunta	
Eurasian beaver (*Castor fiber*)	M52	-29.7	-30.9	-1.2	Satakunta	
Eurasian elk (*Alces alces*)	M57	-31.4	-33.8	-2.4	Oripää	
Eurasian elk (*Alces alces*)	M30	-30.6	-32.2	-1.5	Kuhmo	
Eurasian elk (*Alces alces*)	MM132	-30.4	-32.6	-2.2	Kuhmo	
Eurasian elk (*Alces alces*)	M29	-32.1	-34.0	-2.0	Kuhmo	
Eurasian lynx (*Lynx lynx*)	M122	-32.5	-31.9	0.6	Ilomantsi	
Eurasian lynx (*Lynx lynx*)	MM124	-32.0	-31.8	0.2	Ilomantsi	
Eurasian lynx (*Lynx lynx*)	MM123	-30.9	-31.2	-0.3	n/a	
Eurasian lynx (*Lynx lynx*)	M63	-31.1	-31.1	0.0	n/a	
Grey seal (*Halichoerus grypus*)	M1	-25.4	-25.3	0.1	Isokari, Uusikaupunki, Finnish Archipelago Sea	
Grey seal (*Halichoerus grypus*)	M2	-26.9	-27.1	-0.2	Merikarvia, Sea of Bothnia	
Grey seal (*Halichoerus grypus*)	M3	-24.6	-24.9	-0.3	South Kälö, Korppoo, Finnish Archipelago Sea	
Grey seal (*Halichoerus grypus*)	M51	-24.9	-25.1	-0.2	Pärnu Bay, Gulf of Riga, Estonia	
Grey seal (*Halichoerus grypus*)	MM129	-24.7	-25.3	-0.6	Brändö, Åva, Finnish Archipelago Sea	
Mountain hare (*Lepus timidus*)	M133	-34.1	-34.4	-0.3	North Karelia	
Mountain hare (*Lepus timidus*)	MM121	-33.8	-34.3	-0.5	Kuhmo	
Mountain hare (*Lepus timidus*)	M59	-32.8	-33.3	-0.5	Kuhmo	
Reindeer (*Rangifer tarandus*)	M55	-25.4	-28.2	-2.7	n/a	
Reindeer (*Rangifer tarandus*)	M54	-25.3	-28.2	-2.9	n/a	
Wild boar (*Sus scrofa ferus*)	MM125	-27.9	-27.0	0.8	Ilomantsi	
Wild boar (*Sus scrofa ferus*)	MM131	-28.2	-28.2	0.0	Ilomantsi	

Table 1, cont.

	Lab code	$\delta^{13}C_{16:0}$ (‰)	$\delta^{13}C_{18:0}$ (‰)	$\Delta^{13}C$ (‰)	Place of catchment	Other remarks
Wild forest reindeer (*Rangifer tarandus fennicus*)	M13	-26.4	-29.5	-3.1	Oksakoski, Perho	
Wild forest reindeer (*Rangifer tarandus fennicus*)	M14	-26.1	-27.9	-1.8	Oksakoski, Perho	
Wild forest reindeer (*Rangifer tarandus fennicus*)	M16	-27.0	-28.2	-1.2	Oksakoski, Perho	
Wolf (*Canis lupus*)	MM127	-30.3	-32.8	-2.5	Ilomantsi	
Wolf (*Canis lupus*)	MM128	-28.5	-31.0	-2.5	Tohmajärvi	
Wolf (*Canis lupus*)	MM126	-35.6	-35.0	0.6	Tohmajärvi	Strong human contact?
Wolf (*Canis lupus*)	M67	-28.3	-30.2	-1.9	n/a	
Fish from the Baltic Sea						
Baltic herring (*Clupea harengus membras*)	SF45	-28.1	-26.9	1.2	Sea of Bothnia	
Baltic herring (*Clupea harengus membras*)	SF17	-29.1	-31.8	-2.7	Sea of Bothnia	
Baltic herring (*Clupea harengus membras*)	SF33	-29.0	-28.6	0.4	Sea of Bothnia	
Baltic herring (*Clupea harengus membras*)	SF34	-28.6	-30.8	-2.3	Sea of Bothnia	
Baltic herring (*Clupea harengus membras*)	SF140	-28.4	-27.6	0.9	Bay of Bothnia, Sweden	
Northern pike (*Esox lucius*)	SF37	-32.8	-31.9	0.9	Finnish Archipelago Sea	
Northern pike (*Esox lucius*)	SF16	-24.6	-24.8	-0.2	Rymättylä, Finnish Archipelago Sea	
Northern pike (*Esox lucius*)	SF5	-24.7	-23.7	1.0	Askainen, Finnish Archipelago Sea	
Northern pike (*Esox lucius*)	SF2	-25.5	-24.8	0.7	Askainen, Finnish Archipelago Sea	
Perch (*Perca fluviatilis*)	SF42	-25.3	-23.5	1.8	Sea of Åland	
Perch (*Perca fluviatilis*)	SF43	-25.3	-24.7	0.6	Sea of Åland	
Perch (*Perca fluviatilis*)	SF36	-25.1	-23.8	1.3	Sea of Åland	
Perch (*Perca fluviatilis*)	SF20	-24.6	-24.1	0.5	Sea of Åland	
Pike-perch (*Sander lucioperca*)	SF41	-26.7	-26.2	0.5	Luonnonmaa, Finnish Archipelago Sea	
Pike-perch (*Sander lucioperca*)	SF7	-24.4	-24.2	0.2	Parainen, Finnish Archipelago Sea	
Pike-perch (*Sander lucioperca*)	SF38	-25.1	-24.4	0.7	Rymättylä, Finnish Archipelago Sea	
Atlantic salmon (*Salmo salar*)	SF4	-26.0	-24.6	1.4	Peimari, Finnish Archipelago Sea	
Atlantic salmon (*Salmo salar*)	SF138	-25.3	-24.9	0.4	Askainen, Finnish Archipelago Sea	
Sprat *(Sprattus sprattus*)	SF44	-28.1	-28.6	-0.5	Sea of Bothnia	
Sprat *(Sprattus sprattus*)	SF30	-26.5	-25.9	0.5	Sea of Bothnia	
Sprat *(Sprattus sprattus*)	SF31	-27.9	-27.1	0.8	Sea of Bothnia	
Roach (*Rutilus rutilus*)	SF11	-23.4	-24.9	-1.5	Seili, Finnish Archipelago Sea	
Roach (*Rutilus rutilus*)	SF12	-24.4	-25.0	-0.6	Seili, Finnish Archipelago Sea	
Roach (*Rutilus rutilus*)	SF8	-24.2	-24.0	0.1	Seili, Finnish Archipelago Sea	
Whitefish (*Coregonus lavaretus*)	SF35	-25.2	-25.3	-0.2	Luonnonmaa, Finnish Archipelago Sea	
Whitefish (*Coregonus lavaretus*)	SF39	-27.2	-25.6	1.7	Finnish Archipelago Sea	
Whitefish (*Coregonus lavaretus*)	SF40	-26.2	-25.8	0.4	Luonnonmaa, Finnish Archipelago Sea	
Whitefish (*Coregonus lavaretus*)	SF46	-25.4	-24.5	0.8	Rymättylä, Finnish Archipelago Sea	
Freshwater fish						
Bleak (*Alburnus alburnus*)	LF13	-28.7	-28.0	0.6	n/a	
Bream (*Abramis brama*)	LF3	-35.9	-34.4	1.5	Lake Kellojärvi, Kuhmo	
Bream (*Abramis brama*)	LF144	-36.2	-36.6	-0.4	Lake Niinivesi, Äänekoski	
Bream (*Abramis brama*)	LF152	-37.7	-37.2	0.5	Lake Niinivesi, Äänekoski	
Bream (*Abramis brama*)	LF142	-38.1	-36.6	1.4	Lake Niinivesi, Äänekoski	
Burbot (*Lota lota*)	LF6	-36.4	-34.4	2.0	Lake Kellojärvi, Kuhmo	

Table 1, cont.

	Lab code	$\delta^{13}C_{16:0}$ (‰)	$\delta^{13}C_{18:0}$ (‰)	$\Delta^{13}C$ (‰)	Place of catchment	Other remarks
Ide (*Leuciscus idus*)	PF8	-35.5	-33.9	1.6	Lake Pajalampi, Kuhmo	
Ide (*Leuciscus idus*)	PF5	-34.9	-33.3	1.6	Lake Pajalampi, Kuhmo	
Northern pike (*Esox lucius*)	LF146	-35.3	-35.0	0.4	Lake Pönkälampi, Kuhmo	
Northern pike (*Esox lucius*)	LF4	-35.5	-34.3	1.2	Lake Kellojärvi, Kuhmo	
Northern pike (*Esox lucius*)	LF147	-33.8	-33.4	0.4	Lake Niinivesi, Äänekoski	
Perch (*Perca fluviatilis*)	LF151	-36.4	-35.3	1.1	Lake Niinivesi, Äänekoski	
Perch (*Perca fluviatilis*)	LF148	-37.2	-36.7	0.5	Lake Pönkälampi, Kuhmo	
Perch (*Perca fluviatilis*)	LF32	-35.4	-34.6	0.7	Lake Valkea-Kotinen, Hämeenlinna	
Perch (*Perca fluviatilis*)	LF18	-37.0	-35.4	1.6	Lake Valkea-Kotinen, Hämeenlinna	
Perch (*Perca fluviatilis*)	LF33	-35.0	-34.4	0.7	Lake Valkea-Kotinen, Hämeenlinna	
Perch (*Perca fluviatilis*)	LF143	-34.1	-34.4	-0.2	Lake Murtojärvi, Kuhmo	
Perch (*Perca fluviatilis*)	PF149	-34.1	-34.7	-0.6	Lake Hujakko, Äänekoski	
Perch (*Perca fluviatilis*)	PF2	-37.4	-38.9	-1.6	Lake Pajalampi, Kuhmo	
Perch (*Perca fluviatilis*)	PF1	-37.3	-36.8	0.5	Lake Pajalampi, Kuhmo	
Pike-perch (*Sander lucioperca*)	LF5	-36.6	-35.8	0.8	Lake Kellojärvi, Kuhmo	
Pike-perch (*Sander lucioperca*)	LF9	-32.3	-32.0	0.3	n/a	
Roach (*Rutilus rutilus*)	PF150	-31.5	-34.0	-2.5	Lake Hujakko, Äänekoski	
Roach (*Rutilus rutilus*)	PF3	-36.1	-35.0	1.1	Lake Pajalampi, Kuhmo	
Vendace (*Coregonus albula*)	LF28	-30.0	-31.6	-1.5	Lake Puruvesi	
Vendace (*Coregonus albula*)	LF27	-31.2	-29.9	1.3	Lake Puruvesi	
Vendace (*Coregonus albula*)	LF145	-38.9	-39.9	-1.0	Lake Lentua, Kuhmo	
Vendace (*Coregonus albula*)	LF11	-30.3	-28.9	1.5	Lake Vanajanselkä	

with a GC interface temperature of 250°C and a source temperature of 200°C. Peaks were identified based on their mass spectra using Xcalibur software.

Gas-chromatography-combustion-isotope ratio mass spectrometry (GC-C-IRMS)

Stable carbon isotope values for the fatty acid methyl esters were determined using two different GC-C-IRMS instruments: 1. Agilent 6890 GC coupled to a ThermoFinnigan DeltaPlus XL mass spectrometer via a Finnigan MAT GCCIII interface with a Cu/Ni reactor maintained at 950°C; and 2. Agilent Technologies 7890A GC coupled to an IsoPrime 100 via an IsoPrime GC5 combustion interface with a CuO and silver wool reactor maintained at 850°C. Diluted samples were introduced using a PTV injector in the splitless mode onto a VF-23ms 60 m × 0.32 mm i.d. capillary column coated with cyanopropyl polysiloxane (film thickness, 0.15 μm, Agilent Technologies). The GC oven temperature was programmed from 40°C, following an isothermal hold for 2 min, to 250°C at 10°C/min, followed by an isothermal hold at 250°C for 15 min. Each sample was run in duplicate.

Statistical analyses

Statistical analyses were carried out using SPSS 23.0 software. The Mann-Whitney *U*-test was used to compare the differences between two comparable groups. The Kruskal-Wallis test was used to compare the differences between more than two groups. The differences were reported using *p*-values with Bonferroni correction. The level of significance was set at $p \leq 0.05$. The results of the statistical analyses are summarised in Table 2.

Results

The results from the compound-specific stable carbon isotope ratio analysis of the $C_{16:0}$ and $C_{18:0}$ fatty acids are presented in Fig. 2 and Table 1.

Freshwater species and brackish species

Five grey seal muscle tissue samples from the Baltic Sea were studied. The main fatty acid components of the grey seal muscle tissue were $C_{16:0}$, $C_{16:1}$, $C_{18:1}$, $C_{18:2}$, and $C_{22:6}$. In the fish tissues the main compounds

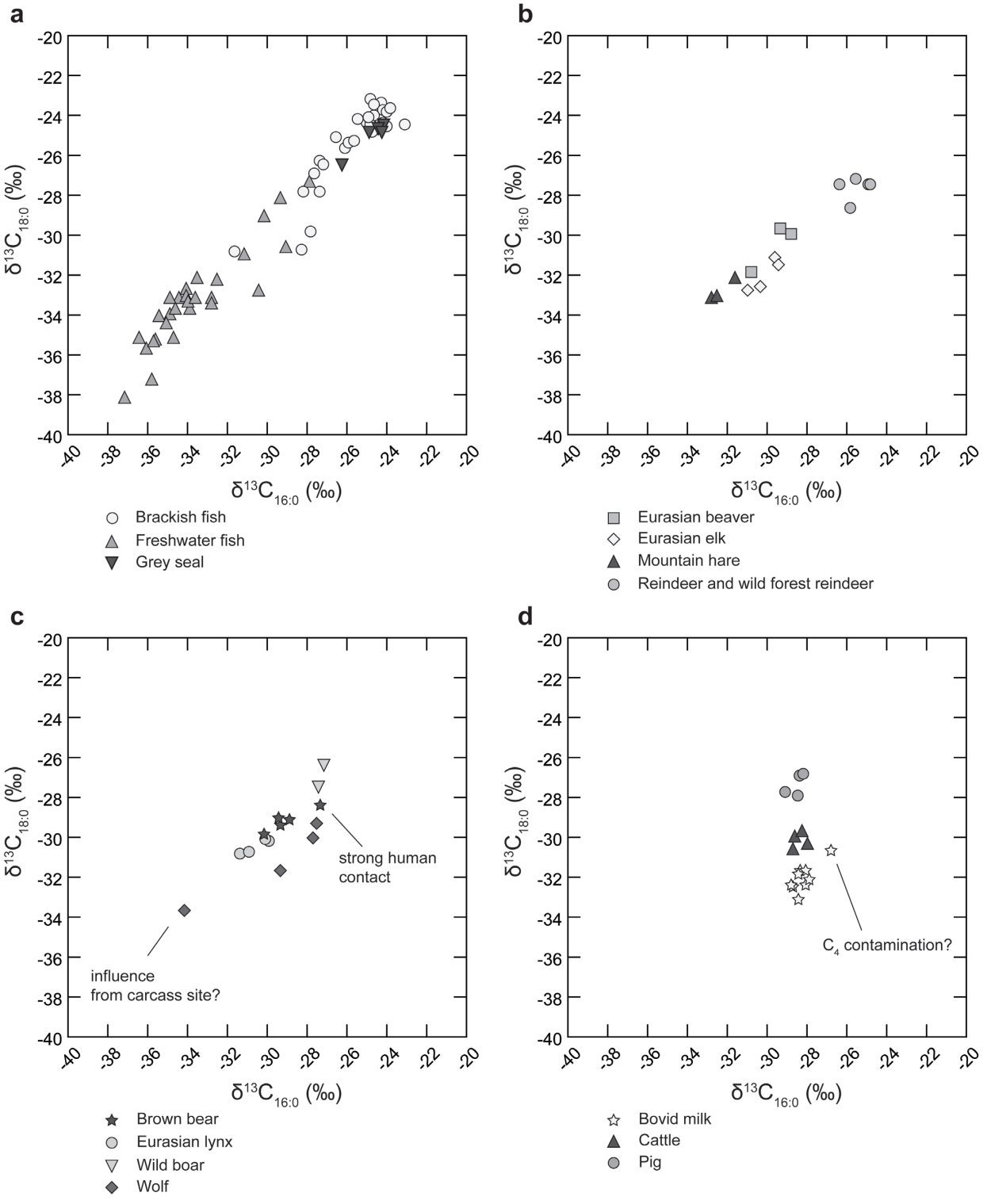

Figure 2. The $\delta^{13}C$ values of the individual *n*-alkanoic-acids $C_{16:0}$ and $C_{18:0}$ of studied muscle tissues and milks: (a) Brackish fish, grey seal, and freshwater fish; (b) Eurasian beaver, mountain hare, and Cervidae; (c) Carnivores and omnivores; and (d) Domesticated pigs, cattle, and bovid milk. Data was plotted using MYSTAT 12, version 12.02.00.

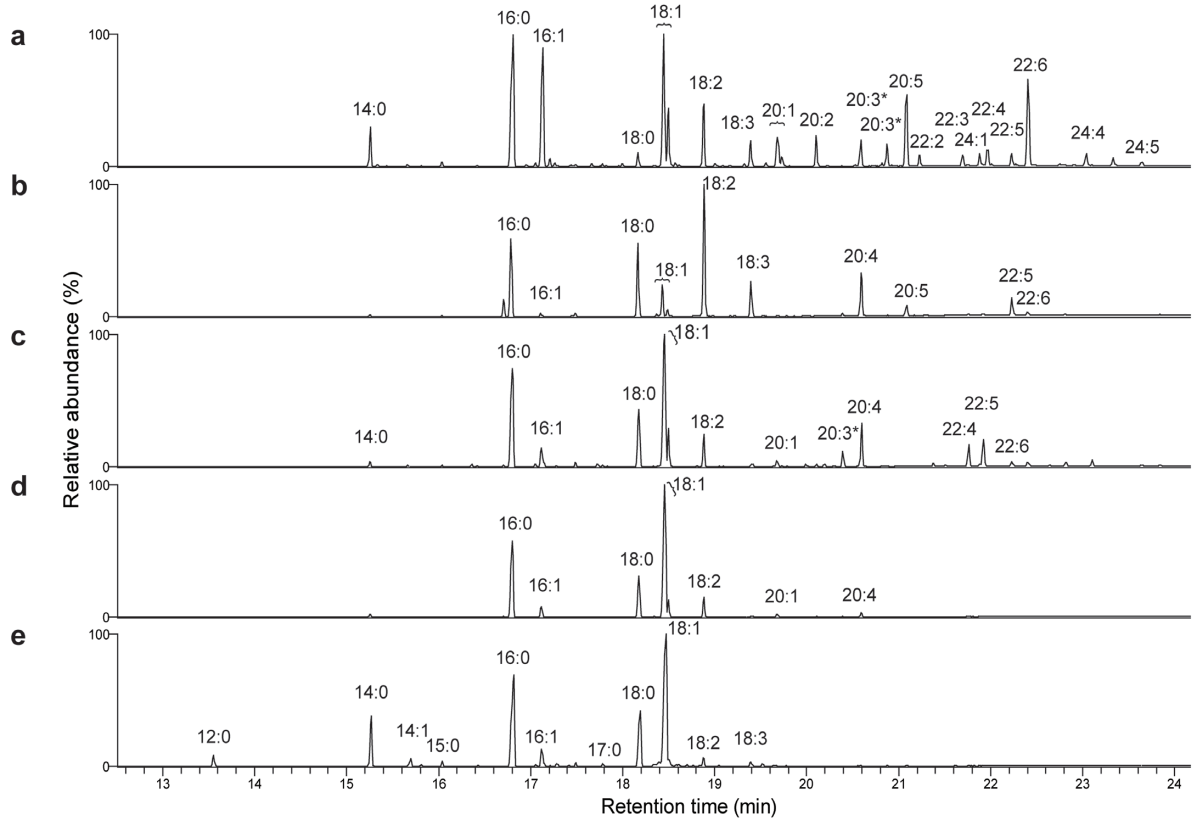

Figure 3. Partial gas chromatograms of (a) Baltic herring, (b) mountain hare, (c) brown bear, (d) domesticated pig, and (e) a milk from indigenous Northern Finn cattle. * Three different isomers of fatty acid $C_{20:3}$ were observed. See Material and methods for full experimental details.

were $C_{16:0}$, $C_{18:1}$, and $C_{22:6}$ (Fig. 3a). These results are in general agreement with previously published compositions (see e.g. Puustinen et al. 1985:60–61; Käkelä et al. 1993:554–556). The $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values for the freshwater fish were significantly different from those of the brackish species (Fig. 2a, Table 2).

Small terrestrial and semi-aquatic mammals

Three muscle tissue samples were analysed from the Eurasian beaver (*Castor fiber*) and the mountain hare (*Lepus timidus)*. The main fatty acid components in the Eurasian beaver muscle tissues were $C_{16:0}$, $C_{16:1}$, $C_{18:1}$, $C_{18:2}$ and $C_{18:3}$, which agrees with previously published compositions (Käkelä & Hyvärinen 1996a:116–117). $C_{18:2}$ was the main fatty acid component in the mountain hare muscle tissue; $C_{16:0}$, $C_{18:0}$, $C_{18:1}$, $C_{18:3}$, $C_{20:3}$, $C_{20:5}$, and $C_{22:5}$ were also significant components (Fig. 3b). The $\delta^{13}C$ values of the Eurasian beaver and mountain hare fats were not significantly different (Fig. 2b, Table 2).

Cervidae

Four Eurasian elk (*Alces alces*) muscle tissue samples, two samples of reindeer, and three samples of wild forest reindeer were studied. There were only minor differences in the fatty acid compositions of all the cervids, with the main fatty acids in Eurasian elk, reindeer, and wild forest reindeer $C_{16:0}$, $C_{18:0}$, $C_{18:1}$, $C_{18:2}$, and $C_{20:4}$, which agrees with previously published compositions (see e.g. Tanhuanpää & Pulliainen 1975:150–151; Wiklund et al. 2001:295). The $\delta^{13}C$ values of the fats from Eurasian elk and reindeer/wild forest reindeer were, however, significantly different (Fig. 2b, Table 2). There was c. 5‰ mean difference in the $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ fatty acids of Eurasian elk and reindeer/wild forest reindeer. Moreover, the $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values of Eurasian elk fats were also significantly different from those of cattle (Table 2).

Table 2. Results of the statistical analyses carried out for the $\delta^{13}C$ and $\Delta^{13}C$ ($\delta^{13}C_{18:0}$ - $\delta^{13}C_{16:0}$) values. Values for marine species from Cramp & Evershed (2014) and values for marine species from coastal Denmark from Craig et al. (2011) and Lucquin et al. (2016). Milk sample ML100 is excluded from the statistical analyses due to possible silage contribution to the diet. The results of the Kruskal-Wallis test are reported using p-values with Bonferroni correction.

		$\delta^{13}C_{16:0}$		$\delta^{13}C_{18:0}$		$\Delta^{13}C$	
Kruskal-Wallis test	**n**	***p***	**corr. *p***	***p***	**corr. *p***	***p***	**corr. *p***
Freshwater fish, brackish species, marine species from UK	87	**< 0.001**		**< 0.001**		0.113	
Brackish species	33						
Freshwater fish	28		**< 0.001**		**< 0.001**		
Freshwater fish	28						
Marine species (UK)	26		**< 0.001**		**< 0.001**		
Brackish species	33						
Marine species (UK)	26		0.413		0.097		
Finnish Archipelago Sea, Sea of Åland, Sea of Bothnia (including Bay of Bothnia)	28	**0.002**		**< 0.001**			
Bay of Bothnia and Sea of Bothnia	8		**0.003**		**0.007**		
Finnish Archipelago Sea	16						
Bay of Bothnia and Sea of Bothnia	8		**0.019**		**0.001**		
Sea of Åland	4						
Finnish Archipelago Sea	16		1.000		0.275		
Sea of Åland	4						
Aquatic fats, milk fats, porcine fats, ruminant fats	88					**< 0.001**	
Milk fats (ML100 not included)	8						1.000
Ruminant fats	13						
Aquatic fats	61						**< 0.001**
Milk fats (ML100 not included)	8						
Milk fats (ML100 not included)	8						**< 0.001**
Porcine fats	6						
Aquatic fats	61						**< 0.001**
Ruminant fats	13						
Porcine fats	6						**0.001**
Ruminant fats	13						
Aquatic fats	61						1.000
Porcine fats	6						
Milk fats, Porcine fats, ruminant fats	27					**< 0.001**	
Milk fats (ML100 not included)	8						**0.010**
Ruminant fats	13						
Milk fats (ML100 not included)	8						**< 0.001**
Porcine fats	6						
Porcine fats	6						**0.046**
Ruminant fats	13						
Freshwater fish, Mountain hare, Eurasian beaver	34	**0.031**		0.068			
Mountain hare	3		0.621				
Freshwater fish	28						
Eurasian beaver	3		**0.047**				
Freshwater fish	28						
Mountain hare	3		1.000				
Eurasian beaver	3						
Aquatic fats, wild terrestrial and semi-aquatic mammals excluding ruminants and wild boar	80					**0.014**	
Eurasian beaver	3						1.000
Wolf	4						
Mountain hare	3						1.000
Eurasian beaver	3						

		$\delta^{13}C_{16:0}$		$\delta^{13}C_{18:0}$		$\Delta^{13}C$	
Kruskal-Wallis test	**n**	***p***	**corr. *p***	***p***	**corr. *p***	***p***	**corr. *p***
Brown bear	5						1.000
European beaver	3						
Eurasian lynx	4						1.000
European beaver	3						
Aquatic	61						0.363
European beaver	3						
Mountain hare	3						1.000
Wolf	4						
Brown bear	5						1.000
Wolf	4						
Eurasian lynx	4						1.000
Wolf	4						
Aquatic	61						0.216
Wolf	4						
Mountain hare	3						1.000
Brown bear	5						
Mountain hare	3						1.000
Eurasian lynx	4						
Aquatic	61						0.883
Mountain hare	3						
Brown bear	5						1.000
Eurasian lynx	4						
Aquatic	61						1.000
Brown bear	5						
Aquatic	61						1.000
Eurasian lynx	4						

		$\delta^{13}C_{16:0}$		$\delta^{13}C_{18:0}$	
Mann-Whitney U-test	**n**	***p***	**Z**	***p***	**Z**
Brackish species	33				
Freshwater fish	28	**< 0.001**	-6.572	**< 0.001**	-6.500
Mountain hare	3				
Europen beaver	3	0.100	-1.964	0.100	-1.964
Eurasian elk	4				
Reindeer/wild forest reindeer	5	**0.016**	-2.449	**0.016**	-2.491
Cattle	4				
Eurasian elk	4	**0.029**	-2.309	**0.029**	-2.309
Brown bear	5				
Eurasian lynx	4	0.063	-1.960	**0.016**	-2.449
Brown bear	5				
Wild boar	2	0.190	-1.549	0.095	-1.936
Cattle	4				
Milk (ML100 not included)	8	0.933	-0.085	**0.004**	-2.727
Cattle	4				
Pig	4	0.886	-0.289	**0.029**	-2.309
Brackish fish	28				
Grey seal	5	0.364	-0.955	0.609	-0.528
Brackish species	33				
Marine species (coastal Denmark)	17	**< 0.001**	-5.706	**< 0.001**	-4.886
Reindeer	2				
Wild forest reindeer	3	0.200	-1.732	1.000	0.000
Cattle	4				
Reindeer/wild forest reindeer	5	**0.016**	-2.449	**0.016**	-2.491

Carnivores and omnivores

In the wild boar (*Sus scrofa ferus*) tissues the main fatty acids were $C_{16:0}$, $C_{18:0}$, $C_{18:1}$, and $C_{18:2}$, which agree with previously published compositions (Koizumi et al. 1991:549). The main fatty acids in the brown bear (*Ursus arctos*) muscle were $C_{16:0}$, $C_{18:0}$, and $C_{18:1}$ (Fig. 3c). One of the bear muscle samples originated from an individual shot in an oat field. This animal had probably been living near human settlements for a substantial period and is likely to have been killed since it was considered dangerous. Nevertheless, it had a similar fatty acid composition to the other samples from the same species. In the Eurasian lynx (*Lynx lynx*) muscle tissue the main fatty acids were $C_{16:0}$, $C_{18:0}$, $C_{18:2}$, and $C_{20:4}$. The main fatty acids in the wolf (*Canis lupus*) muscle were $C_{16:0}$, $C_{18:0}$, $C_{18:1}$, and $C_{18:2}$, agreeing with previously published compositions (Käkelä & Hyvärinen 1996b:509). The $\delta^{13}C_{16:0}$ values of the Eurasian lynx and brown bear muscle tissues were not significantly different, but the $\delta^{13}C_{18:0}$ values were. The wild boar $\delta^{13}C_{18:0}$ values were not significantly different from other studied wild omnivores, i.e. brown bear (Fig. 2c, Table 2).

Domesticated animals and milk

Nine samples of cow and goat's (*Capra hircus)* milk and four muscle tissue samples from both cattle (*Bos taurus*) and pigs (*Sus scrofa domesticus*) were analysed. The main fatty acid in all these domesticated animal tissues and milk was $C_{18:1}$ (Fig. 3d–e). In the milk samples the next most abundant components were even carbon-numbered fatty acids $C_{12:0}$, $C_{14:0}$, $C_{16:0}$, and $C_{18:0}$. In the muscle tissues, $C_{16:0}$, $C_{18:0}$, and $C_{18:2}$ were significant components. These results agree with previously published compositions (e.g. Koizumi et al. 1991:547; Vanhatalo et al. 2007:861–862). The $\delta^{13}C_{16:0}$ values of cattle and milk, and cattle and pigs were not significantly different; however, the values for $\delta^{13}C_{18:0}$ were significantly different (Fig. 2d, Table 2).

Discussion

Brackish fats

Fish are usually more ^{13}C-depleted in freshwater than in brackish environments (Kiljunen et al. 2006:1218). In Finland the bedrock and till are mainly noncarbonated (Henriksen et al. 1998:83; Rasilainen et al. 2008). In addition, in Finnish freshwater environment total inorganic carbon (TIC) mainly derives from catchment weathering and bicarbonate ion is the main form of dissolved inorganic carbon (DIC) in freshwater systems (Rantakari & Kortelainen 2008). In addition, DIC in freshwater derives also from decomposition of terrestrial detritus that is ^{13}C-depleted. Thus, the ecological environment of freshwater systems create conditions where the $\delta^{13}C$ values are more depleted compared to marine ecosystems (Boutton 1991). Indeed, in this investigation, fish from freshwater were found to have significantly more depleted $\delta^{13}C$ values than fish and grey seal samples from the Baltic Sea. Moreover, the grey seal $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ fatty acids $\delta^{13}C$ values were not significantly different to those of the brackish fish (Table 2). This result was expected since Baltic herring (*Clupea harengus membras*) is the principal component of the grey seal's diet (Lundström et al. 2013:184).

The $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ fatty acids of the freshwater species were more depleted and significantly different to those of the marine species (both fish and mammals) from the UK (Cramp & Evershed 2014). However, brackish and marine species did not have significantly different $\delta^{13}C$ values (Table 2). No significant difference was observed in the $\Delta^{13}C$ values of freshwater fish, brackish species, and marine species from the UK. The lack of difference between $\Delta^{13}C$ values is expected, as the $\Delta^{13}C$ proxy is designed to remove ecosystem differences and accentuate metabolic differences between the animals from which the fatty acids derive. Thus, the $\Delta^{13}C$ proxy cannot distinguish between anadromous, littoral, or pelagic fish as they are all metabolically similar. The raw $\delta^{13}C$ values, particularly $\delta^{13}C_{16:0}$, characterise ecosystem affiliations.

Overall, fish from different basins of the Baltic Sea exhibit significantly different $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values. The Bonferroni corrected *p*-values, however, are not significantly different for $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ between the Finnish Archipelago Sea and the Sea of Åland (Table 2). In addition, the marine fats obtained from the Belt Sea on the coast of Denmark (Craig et al. 2011; Lucquin et al. 2016), displayed significantly different $\delta^{13}C$ values compared to those from the brackish species investigated herein. The more depleted $\delta^{13}C$ values for the fish $C_{16:0}$ and $C_{18:0}$ fatty acids from the Sea of Bothnia correlate with the salinity gradient, with salinity being lower in the Sea of Bothnia than in the Finnish Archipelago Sea and in the Sea of Åland (Fig. 1). Due to lime rich soils, alkalinity and DIC content of the riverine inflow to the Baltic Sea is higher in the Eastern parts of the Europe compared to the inflow coming from Scandinavia. As a result, the Gulf of Finland and the Sea of Bothnia exhibit low DIC and alkalinity (Thomas & Schneider 1999:57).

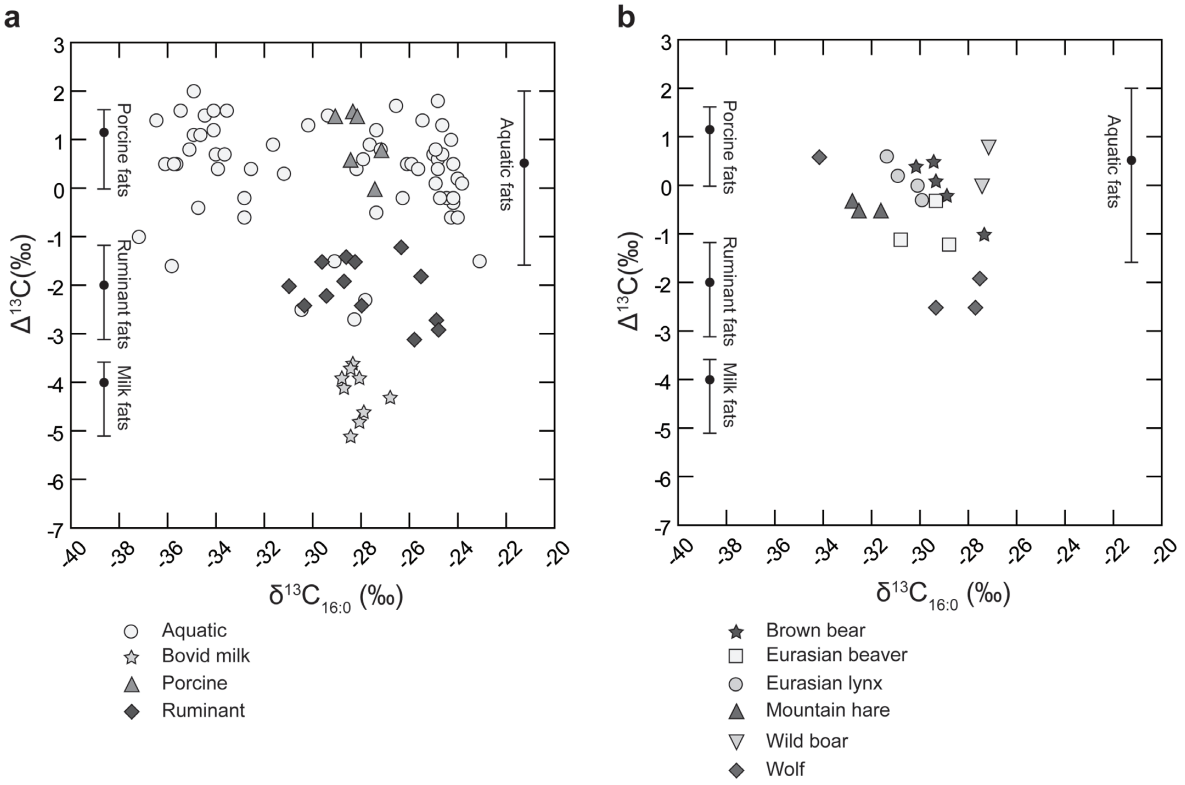

Figure 4. Distribution of the $\Delta^{13}C$ ($\delta^{13}C_{18:0} - \delta^{13}C_{16:0}$) values of fatty acids of studied muscle tissue and milks: (a) Aquatic fats (freshwater fish, brackish fish, and grey seal), bovid milk, porcine fats (pig and wild boar), and ruminant fats (cattle, Eurasian elk, reindeer, and wild forest reindeer); (b) the $\Delta^{13}C$ values of wild terrestrial and semi-aquatic mammal fats, excluding ruminant animals. The whiskers of aquatic, porcine, ruminant, and milk fats show the range of $\Delta^{13}C$ values when outliers are excluded. The circles in whiskers indicate the median of $\Delta^{13}C$ values. Data were plotted using MYSTAT 12, version 12.02.00.

In addition, the $\delta^{13}C$ values of Baltic Sea sediments are lower in coastal areas compared with those of basin sediments (Voss et al. 2000:292, 296). The significant difference in the $\delta^{13}C$ values between the Danish coast and the Northern Baltic Sea reflects the different ecological environment, as the riverine freshwater supply is higher in the northern parts of the basin.

Identification of terrestrial and semi-aquatic mammal fats from aquatic fats

The $\Delta^{13}C$ values have helped to determine domestic ruminant and non-ruminant animal exploitation based on animal fat residues preserved in archaeological pottery (Copley et al. 2003). Likewise, this study confirms that the fats of different groups of animals, namely, aquatic, porcine, ruminant, and bovid milk fats, exhibit significantly different $\Delta^{13}C$ values (Fig. 4a). However, no significant difference was observed in the Bonferroni corrected *p*-values between aquatic species and porcine fats, and milk and the ruminant fats (corrected values appear in Table 2). Nonetheless, when comparing $\Delta^{13}C$ values of only milk, ruminant, and porcine fats the difference was significant between all of the groups (Table 2).

The $\delta^{13}C_{16:0}$ values of the Eurasian beaver, mountain hare, and freshwater fish were significantly different. However, the specific comparisons between these groups indicated a significant difference only existed in the $\delta^{13}C_{16:0}$ values between the freshwater fish and Eurasian beaver (Table 2). In addition, even though the overall effect was significant, the wild terrestrial and semi-aquatic mammals displayed similar $\Delta^{13}C$ values to those of the aquatic organisms (Table 2, Fig. 4b). Thus, based on this study, terrestrial and semi-aquatic mammals, excluding ruminant animals, cannot be distinguished from freshwater fish just based on the $\Delta^{13}C$ proxy. This confirms validity of the original design of the proxy, which was to classify fats based on the metabolic characteristics of the organisms from which they derive. To circumvent the problem of separating the fats of terrestrial and aquatic organisms in the organic residue analysis of archaeological pottery ω-(*o*-alkylphenyl)alkanoic acids (APAAs), vicinal dihydroxy acids, and isoprenoid fatty acid biomarkers are used. These compounds provide an independent,

complementary approach to characterising the processing of aquatic commodities (e.g. Copley et al. 2004; Hansel et al. 2004; 2011; Evershed et al. 2008; Hansel & Evershed 2009; Cramp & Evershed 2014). Aquatic biomarkers, particularly APAAs, absorbed in archaeological pottery are highly resistant to degradation (Cramp & Evershed 2014). Moreover, the formation of APAAs is contingent on the exposure of fats containing the mono-, di-, and triunsaturated fatty acids to temperatures approaching 300°C (Hansel et al. 2004). APAAs are known to derive also from fats other than aquatic origin. However, it has been demonstrated that if APAAs of carbon length C_{18}, C_{20}, and particularly C_{22} are detected, they can be considered as being of marine rather than terrestrial origin (Evershed et al. 2008). In archaeological vessels aquatic commodities could have been processed at low temperatures, i.e. drying, smoking, and fermenting. Thus, the absence of APAAs does not necessarily mean that aquatic products had not been processed in the vessel, and careful use of vicinal dihydroxy acids and isoprenoid fatty acids, together with the raw $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$, are recommended.

Effect of foddering and the identification of wild and semi-domesticated animals

Reindeer are semi-domesticated animals and in Finland the additional feeding (foddering) of reindeer has become increasingly common since the 1960s. Currently, only a small portion of these animals overwinter without fodder (Kemppainen et al. 2003:20). Thus, even in the case of a freely grazing animal, the purity of the C_3 diet cannot be guaranteed and even wild animals might not have consumed their typical staple diet. Wild carnivores can gain a significant amount of their nutrition from carcasses that are used to attract wild animals to feeding sites for the purposes of photography, simple wildlife observation, or hunting. Moreover, in Finland, the remains of fish guts are a permissible material for use at such carcass sites (Pohja-Mykrä & Kurki 2009:25–26). The $\delta^{13}C$ values of the fatty acids recorded for the wolf muscle sample (MM126) are a possible indication of such a carcass feeding phenomenon. This sample plots differently compared to other wolves, near to freshwater fish (Fig. 2). In Finland, the most common prey for wolves is the Eurasian elk, accounting 88–96% of the staple diet for wolves, with fish forming generally < 0.5% of their diet (Gade-Jørgensen & Stagegaard 2000:541). Thus, it is possible that the wolf had scavenged fish from carcass sites as readily accessible food sources.

Differences in the diets of various cervids are recorded in the $\delta^{13}C$ values of the fatty acids. As stated earlier, reindeer can receive additional feed beyond their wild diet, but in this study, the $\delta^{13}C$ values of reindeer and wild forest reindeer were not significantly different (Table 2). The $\delta^{13}C$ values of the fatty acids for caribou (*Rangifer tarandus*) from Canada (Taché & Craig 2015:181) are comparable with those reported here. The difference between the $\delta^{13}C$ values of the fats of reindeer/wild forest reindeer and the fats of Eurasian elk can possibly be explained by the dissimilar diets of these animals. The diet of freely grazing reindeer varies throughout the year, but nearly two-thirds of the vegetable matter consumed is lichens. Critically, the winter survival of most reindeer populations depends on the availability of reindeer lichens (*Cladonia* spp., Helle & Aspi 1983:338; Helle et al. 1990; Nieminen & Heiskari 1989). The diet of Eurasian elk constitutes mainly the leaves of deciduous trees and pine (*Pinus sylvestris*, Hörnberg 2001). The bulk $\delta^{13}C$ values for grey reindeer lichen (*Cladina rangiferina*) and green reindeer lichen (*Cladina mitis*) are reported to vary between –26.0 and –23.8‰ (Teeri 1981:83; Brooks et al. 1997:304; Oelbermann & Schiff 2008:2041). Deciduous tree leaves have $\delta^{13}C$ values ranging from –29.8 to –27.2‰ (Brooks et al. 1997:304). Thus, the consumption of lichens by reindeer account for the significant difference in the $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values for the reindeer/wild forest reindeer and the Eurasian elk.

As stated above reindeer/wild forest reindeer can be distinguished from Eurasian elk based on their fatty acids $\delta^{13}C$ values and, significantly, they can also be differentiated from cattle. Thus, when studying archaeological pottery from areas where reindeer or wild forest reindeer would have formed an important part of the human diet, both $\Delta^{13}C$ and $\delta^{13}C$ values could be used to identify the processing of the tissues of these different ruminants through fats preserved in organic residues. The $\Delta^{13}C$ value can reveal the presence of ruminant fats in the archaeological pottery, while enriched $\delta^{13}C$ values indicate the ruminant fats originating from reindeer or wild forest reindeer.

Effect of additional feeding on domesticated animals

All the farm animal and milk samples studied here were sourced from organic farms in an effort to ensure the animals were raised on a C_3 diet. Unfortunately, it is very difficult to know the exact diet of farmed animals without having direct contact with the farmer. In some cases where the organic meat samples were

purchased from supermarkets, it was difficult to confirm the original farm where the animals were raised, and establish if they had been fed a pure C_3 diet. The contribution of C_4 plants to the diet of farm animals can have an effect on the carbon isotope values of their fats (e.g. Roffet-Salque et al. 2016:3, 6). Additionally, cereal can constitute a major part of concentrated cattle feed; in Finland, as much as 50% of a cow's diet can be concentrated feed during the first three months of lactation (Evira 2016). Thus, milk fats especially could be affected by an impure C_3 diet or diets which incorporate concentrates. It is possible that one of the studied milk samples, ML100, was affected by an impure C_3 diet, as the $\delta^{13}C$ values differ from all the other milk fat samples investigated. Recent work has also shown the impacts of ensiled animal feed on the carbon isotope compositions of cattle fatty acids (Roffet-Salque et al. 2016); ensiling is a modern farming innovation that would not have been practised in prehistoric times. Furthermore, woodland pasturing is practised in Finland, although this would not have had a significant effect on the $\delta^{13}C$ values of the fatty acids in cattle carcass and milk fats compared to open grazing (Copley et al. 2003).

Cereals are also the main constituent of pig feed; in 2005 the average pig feed contained c. 80% cereal. Furthermore, pigs can be fed fish meal (Karhapää et al. 2005; Evira 2014). Nevertheless, in farms which used fish or fish meal in the feed, the percentage of fish/fish meal would have been rather low, probably only c. 2–7% of the entire feed (Karhapää et al. 2005). This additional feeding would have only a minor effect on the carbon isotope values of the fatty acids of animals receiving such diets. The only way to ensure the purity of these animals' diets would be to conduct a feeding study (Webb et al. 2017). Nevertheless, when using modern fats as a comparison to support archaeological investigations, it must be remembered that even the diet of prehistoric domesticated animals may have contained something that people today would consider as unexpected or unusual for those animals. For example, based on Finnish historical sources, sheep were given lichen as winter fodder (Kortesalmi 1975:370). As shown for reindeer/wild forest reindeer, lichen alters their fatty acid $\delta^{13}C$ values compared to the values recorded for other ruminant animals. It is for this reason that the $\Delta^{13}C$ proxy is used to remove environmental effects to allow categorisation of fats based on the metabolism of the animals from which they derived. However, caution must still be exercised where highly specialised or unusual diets are concerned. For example, in Iceland, animal fodder was supplemented with a range of animal products, including fish, fish offal and seal fat (Amorosi et al. 2013).

Conclusion

The $\delta^{13}C$ values of fatty acid components of organic residues absorbed in the walls of archaeological pottery are increasingly being used in archaeological investigations of human diet and animal exploitation. When identifying the source of ancient fat preserved in archaeological vessels, a comparison of $\delta^{13}C$ values and $\Delta^{13}C$ values of modern fat values is vital. When comparing modern stable carbon isotope values to archaeological values, it should be noted that the values of modern fats, even from wild animals, are likely to be affected by modern human impacts. Thus, the purity or representativeness of these modern fats requires critical consideration.

However, by careful selection, this study has provided $\delta^{13}C$ values that can be used when studying Northern hunter-gatherer-fisher societies and early farmers. The stable carbon isotope data reported in this paper demonstrate the importance of using modern reference fat data from similar ecological conditions to those of the studied archaeological site, especially where wild fauna are known to have contributed significantly to human diet. We also show that even though reindeer and wild forest reindeer share a similar $\Delta^{13}C$ proxy with other ruminant animals, due to their common digestive physiologies, they can be distinguished from other ruminants based on their more enriched $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values. This difference in $\delta^{13}C$ values is likely due to the lichen-rich diet of reindeer and wild forest reindeer.

In addition, it should be stressed that the Baltic Sea is a vast basin and to be able to create a comprehensive database that covers the whole basin and its catchment, more fish and mammal samples need to be gathered from the southern and most northern regions of the sea, as well as the coastal zones, to confirm the trends observed herein.

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